

# Greenhouse Gas Emission by Static Chamber and Eddy Flux Methods

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## Abstract

Wetlands, including irrigated rice (*Oryza sativa* L.) fields, have soil conditions suitable for production of all major greenhouse gases (CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O). Measurement of gas emission to the atmosphere has largely depended on chamber methods, especially static chamber (commonly called closed-chamber) methods. In addition, flow-through (dynamic) chamber methods are also used, where the increase in gas concentration in an effluent stream is measured and compared with that in the influent stream. This method requires an adequate measurement of flow rate and gas concentration in the flow stream. With the advantage of representing a larger area in the field, tower-based micrometeorological approaches have been developed. Micrometeorological methods can take several forms: profile methods, eddy accumulation methods, and eddy covariance methods. Profile methods, which are based on flux-gradient similarity relationships and utilize measurements at different heights above the ground, are not covered in this chapter. Based on the principles of eddy motions, eddy accumulation and eddy covariance methods are preferred because they are a direct measure of fluxes and require no additional empirical constants. Eddy-based methods are reliant on the ability to capture updrafts and downdrafts of gas concentrations in the atmosphere. This requires the ability to measure all three vector components of wind speed at a high measurement frequency. Modern methods have advanced in available instrumentation and are now able to take advantage of three-dimensional sonic anemometers and small-scale, field-deployable gas analyzers.

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Wetlands are atmospheric sources of  $\text{CH}_4$ ,  $\text{CO}_2$ , and  $\text{N}_2\text{O}$ , which contribute to the Earth's greenhouse effect. Wetlands, including irrigated rice fields, have soil conditions suitable for both  $\text{CH}_4$  and  $\text{N}_2\text{O}$  production. The production and emission of these greenhouse gases is greatly affected by the physical, chemical, and biological properties of the soils. Measurement of greenhouse gas emission to the atmosphere has largely depended on chamber methods, especially static chamber methods that are described in detail in this chapter. In addition to the static-chamber (also commonly called closed-chamber) methods there are also flow-through (dynamic) chamber methods where the increase in gas concentration in an effluent stream is measured and the flux rate of the gas from the soil is calculated from the increase in the gas concentration in the outlet stream over that in the influent stream (DeLaune et al., 2002; Rolston, 1986). This method requires an adequate measurement of flow rate and gas concentration in the flow stream.

With the advantage of representing a larger area in the field, tower-based micrometeorological approaches have also been used to measure the flux of greenhouse gases. Micrometeorological methods can take several forms: profile methods, eddy accumulation methods, and eddy covariance methods. Profile methods are based on flux-gradient similarity relationships and utilize measurements at different heights above the ground. While these methods are the most technologically and economically accessible, in recent years they have been increasingly replaced by the more sophisticated eddy flux based techniques. Based on the principles of eddy motions, eddy accumulation and eddy covariance methods are preferred because they are a direct measure of fluxes and require no additional empirical constants (Foken, 2008). Only the eddy-based techniques are addressed here. For a description of the profile-based methods, see Foken (2008) and Stull (2001). It should also be noted that the terms *eddy covariance* and *eddy correlation* are often seen interchangeably in the literature; it is preferable, however, to use the term *eddy covariance* to avoid confusion with the calculation of fluxes from other methods based on mathematical correlations (Foken, 2008). Eddy-based methods are reliant on the ability to capture updrafts and downdrafts of gas concentrations in the atmosphere. This requires the ability to measure all three vector components of wind speed at a high measurement frequency. Modern methods have advanced in available instrumentation and are now able to take advantage of three-dimensional sonic anemometers and small-scale, field-deployable gas analyzers.

### STATIC CHAMBER MEASUREMENT

The static (closed) chamber method is the system generally used in determining gaseous flux to the atmosphere from wetlands. The closed-chamber method described by Rolston (1986) is the most common method used for measuring gas exchange between the soil and the atmosphere. Because of its low cost and simplicity of application, it is used extensively in various ecosystems including wetlands, especially in areas where a power supply is not available. Flux measurements can be taken multiple times during the year for estimating seasonal or annual flux. Gas chambers consist of base units (25 by 25 by 15 cm) constructed

of clear, 0.64-cm-thick acrylic plastic (Lindau and DeLaune, 1991) (Fig. 22-1). Each base unit is placed at approximately the 10-cm depth in the soil. Water is used to seal the tops (25 by 25 by 25 cm) to the chamber bases. The easily removable top ensures that soil-entrapped gas bubbles (e.g., CH<sub>4</sub>) are not disturbed and released. A rubber septum is used as a sampling port, and a thermometer is installed to monitor the temperature inside the chamber. The method is adequate for measuring gas fluxes for a short period of time following placement of the closed chamber over the base unit. Gas must be mixed so that a concentration gradient does not occur. Mixing is normally accomplished by diffusion in small chambers. A small fan may be required to ensure mixing in large static chambers.

Gas samples are taken with a 15-mL plastic syringe and a 20-gauge stainless steel hypodermic needle. Samples are collected at 0, 10, 20, and 30 min (or a suitable time interval to have a linear buildup of the concentration of the gas being measured) after chamber top placement. Fifteen milliliters of the sample gas is injected into a Vacutainer (13 mL). The overpressure created will ensure that atmospheric gases will not contaminate the sample gases. Silicone sealant is used to seal the injection hole in the Vacutainer's rubber septum. The CH<sub>4</sub>, CO<sub>2</sub>, or N<sub>2</sub>O concentrations of the gas samples can be measured on a gas chromatograph (GC). The reflux of gases from the soil surface is calculated from the data obtained from the GC. Seasonal and diurnal measurements should be conducted. Adequate replications should be used at each wetland location.

The flux of N<sub>2</sub>O (or other gas) from the wetland soil surface can be estimated using the closed-chamber equation (Rolston, 1986):

$$f(\text{gas}) = \frac{V \Delta C}{A \Delta t}$$

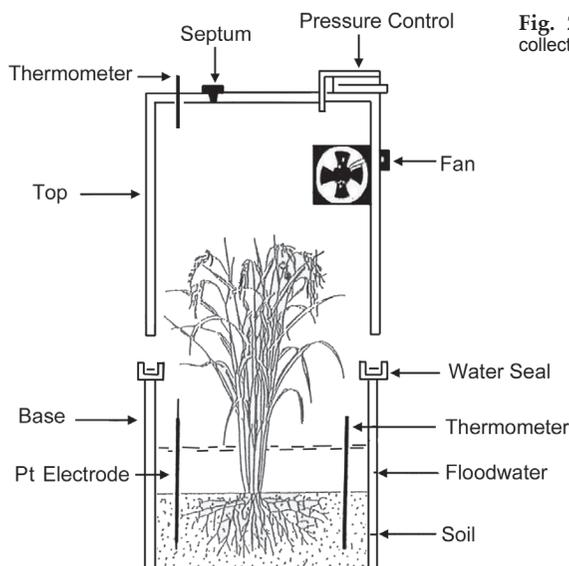


Fig. 22-1. Static (closed) chamber for collection of evolved soil gases.

where  $f$  is the  $N_2O$  gas flux ( $g \text{ gas m}^{-2} \text{ s}^{-1}$ ),  $V$  is the volume of chamber headspace ( $m^3$  gas volume),  $A$  is the soil surface area, and  $\Delta C/\Delta t$  the change in  $N_2O$  gas concentration per unit of time within the chamber (the slope in Fig. 22-2; in  $mg \text{ gas m}^{-3} \text{ s}^{-1}$ ).

A sample calculation of  $N_2O$  flux starts with

$$f(N_2O) = \frac{V \Delta C}{A \Delta t} = \frac{H \Delta C}{\Delta t}$$

where  $H$  is the height of the enclosed chamber headspace (above the water table or soil surface).

In this example (Fig. 22-3), the total height of the chamber is 0.3 m (0.05 m from the base of the chamber + 0.25 m from the top of the chamber). The  $N_2O$  concentration change during 1800 s ( $30 \text{ min} \times 60 \text{ s min}^{-1} = 1800 \text{ s}$ ) is  $0.34 \text{ mg m}^{-3}$  (from  $0.54 \text{ mg m}^{-3}$  at 0 time linearly increased to  $0.88 \text{ mg m}^{-3}$  at the end of 30 min of enclosure).

The flux rate is

$$f(N_2O) = \frac{0.3 \text{ m} \times 0.34 \times 10^{-3} \text{ g m}^{-3}}{1800 \text{ s}} = 0.057 \times 10^{-6} \text{ g N}_2\text{O m}^{-2} \text{ s}^{-1}$$

### Flame Ionization Detection of Carbon Dioxide and Methane

The catalytic conversion-flame ionization technique initially was developed to detect a microliter per liter level of carbon oxides by gas chromatography (Colket et al., 1974). It improved and has been demonstrated to be a very accurate trace analysis technique for trace quantities of carbon oxides.

After separation on the gas chromatograph column, the eluting carbon oxides are hydrogenated over a pure Ni catalyst, and the resulting  $CH_4$  is subsequently detected by a flame ionization detector (FID). The Ni catalyst has a limited life, which could be increased by the introduction of valve venting and the use of a splitter column.

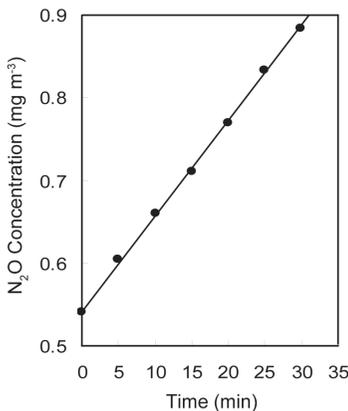


Fig. 22-2. Concentration of  $N_2O$  within a closed chamber as a function of time.

**Equipment**

- Gas chromatograph equipped with an FID, Varian Model 3700 or equivalent
- 46 by 46 cm (18 by 1/8 inch) stainless steel column packed with Porapak Q
- Metal injector insert with a  $\text{Ni}(\text{NO}_3)_2$  catalyst (catalytic convertor)
- Varian CDS III integrator or equivalent
- 0.25-mL sample loop

**Reagents**

- Standard gas cylinders for  $\text{CO}_2$  and  $\text{CH}_4$
- $\text{H}_2$  gas to carry the catalyst. (The  $\text{H}_2$  necessary for the reduction reaction is provided either by using a  $\text{H}_2$  carrier gas or by teeing it in between the column exhaust and the catalyst tube inlet. In the latter case, the usual carrier gas may be used.)
- $\text{N}_2$  carrier gas set at a flow rate of  $20 \text{ mL min}^{-1}$

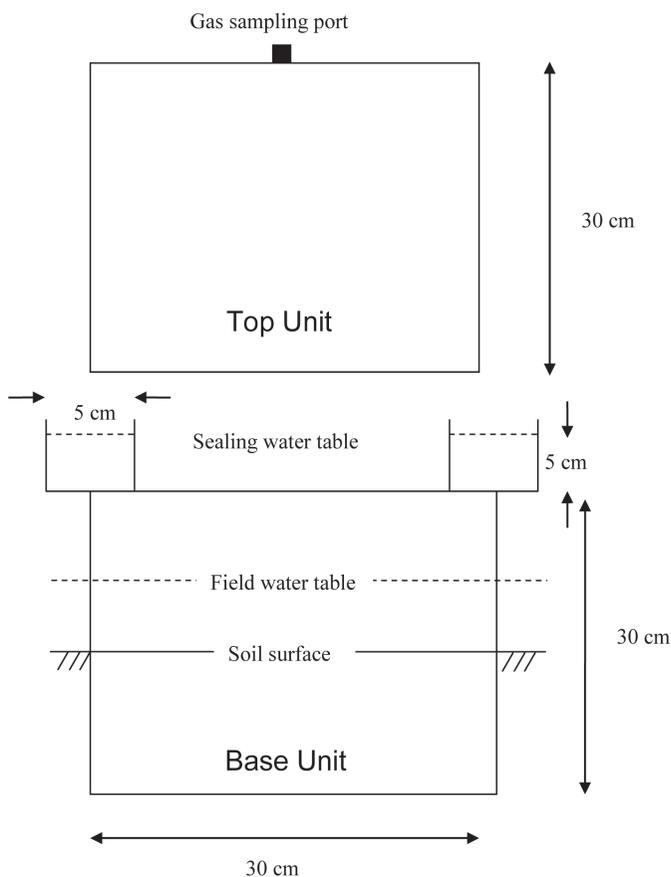


Fig. 22-3. Side view of a static chamber.

### Operating Conditions

These conditions are dependent on instrument requirements and the specific needs of the analyst:

- Detector temperature: 380°C
- Catalytic convertor temperature: 400°C
- Injection port temperature: 40°C
- Column temperature: 50°C

### Procedure

Air samples are collected in conventional glass syringes greased with Apeizon N and sealed with Pharmaseal Teflon three-way stopcocks. The samples should be analyzed within 24 h of their collection. Peak areas are converted into gas concentration ( $\mu\text{L L}^{-1}$ ) and recorded.

### Nitrous Oxide Measurements by Gas Chromatograph

It has been suggested that  $\text{N}_2\text{O}$  production may contribute to destruction of the atmospheric ozone layer. It may also contribute to the atmospheric greenhouse effect by influencing the radioactive budget of the troposphere. The following technique is a good aid for measuring its emissions.

### Equipment

- Varian 3700 gas chromatograph equipped with a  $^{63}\text{Ni}$  electron capture detector and a 0.5-mL sample loop or equivalent
- Chromosorb 106 column 80/100 mesh, 250 cm by 0.2 cm i.d. or equivalent
- Varian CDS III computing integrator or equivalent
- $\text{N}_2\text{O}$  calibration standards (Scott Specialty Gases)
- Carrier gas: 95% Ar with 5%  $\text{CH}_4$
- Glass syringes (10 mL) greased with Apeizon N and sealed with Pharmaseal Teflon three-way stopcocks or equivalent

### Procedure

Gas chromatograph operating parameters vary greatly from instrument to instrument and among testing procedures. The operating conditions for performance of this test on a Varian 3700 gas chromatograph are as follows:

- Detector temperature: 380°C
- Column temperature: 40°C
- Carrier gas flow rate: 30  $\text{mL min}^{-1}$

### EDDY COVARIANCE MEASUREMENT

The eddy covariance technique is most often used for  $\text{CO}_2$  and water vapor fluxes, and there are standard measurement systems and packages available for these gases. The technique is widely applied in micrometeorology over a number of

surfaces (Baldocchi, 2003). For a good review of a number of studies over wetland sites specifically, see Lund et al. (2010). More recently, due to the development of new commercially available sensors, CH<sub>4</sub> and other greenhouse gases have also been measured via eddy covariance. (e.g., Hargreaves et al., 2001; Rinne et al., 2007). Despite the readily available equipment, the eddy covariance method still requires a detailed understanding of atmospheric turbulence, and a variety of corrections is necessary to report accurate fluxes (Foken, 2008).

To achieve a flux measurement, two values are required: a measure of the vertical wind component (i.e., the vector of the wind perpendicular to the ground surface) and a measure of the gas concentration. These two parameters must be measured at a high frequency to determine the flux. The flux is then found as the covariance between the vertical wind velocity ( $w$ ) and the scalar ( $q$ ) of interest. Mathematically this is a measure of how the two variables change together, and it is a proxy for vertical flux of the scalar. The equation is based on the statistical definition of the covariance:

$$\overline{w'q'} = \frac{1}{N-1} \sum_{k=0}^{N-1} [(w_k - \bar{w}_k)(q_k - \bar{q}_k)]$$

where the prime represents the deviation from the mean, also called the *turbulent component*, and the overbar indicates the mean. To determine fluxes from this equation, it is necessary to measure the turbulent components of both the vertical wind speed ( $w$ ) and the scalar of interest. The measurements must be made at a high repetition rate (10–20 Hz) to record the high-frequency end of the turbulent spectrum (Foken, 2008).

The vertical wind component can be determined with a three-dimensional sonic anemometer. Three-dimensional sonic anemometers are available from a variety of manufacturers, including Campbell Scientific, Applied Technologies, Gill Instruments, Metek, and R.M. Young. When choosing a sonic anemometer, it is important to consider path length, temporal resolution, noise specifications, and the maximum range of wind speeds that can be measured. Any physical restrictions to the setup should also be considered. Finally, it is desirable for the sonic anemometer and gas sensor, as well as any ancillary measurements, to be recorded by the same datalogging system, so instrument compatibility should also be a factor in choosing a model. Depending on the scalar, a variety of instruments is available. These include both open- and closed-path sensors. Open-path sensors sample the air that passes through them, while closed-path sensors generally suck in a volume of air that is then analyzed in an enclosed chamber. Open-path analyzers are generally preferred but are not available for all systems. If a closed-path sensor is used, there is additional need for data corrections to ensure that the concentration measurement and wind measurement are timed properly and that no filtering took place in the sample tube (Foken, 2008). Some of the most widely used sensors available today are listed in Table 22-1. This does not represent a complete list because many techniques for measuring gas concentrations exist and many researchers develop or adapt their own methods to do so.

**Table 22-1.** Common gas analyzers available for use in eddy covariance measurements of greenhouse gases. An example study that has used the instrument in wetland environments is provided. This is not a comprehensive list of the use of these instruments for eddy covariance measurements over other land covers, but represents the most common instruments found in the wetlands related literature.

Gas	Instrument	Manufacturer	Type	Example study
CO <sub>2</sub> , H <sub>2</sub> O	Li-7500†	Li-Cor	open path	Glenn et al. (2006)
CO <sub>2</sub> , H <sub>2</sub> O	Li-7000†‡	Li-Cor	closed path	Syed et al. (2006)
H <sub>2</sub> O	KH20 krypton hygrometer†	Campbell Scientific	open path	Dahm et al. (2002)
CH <sub>4</sub>	tunable diode laser spectrometer	Aerodyne Research	closed path	Hargreaves et al. (2001)
Configurable for two or three of: N <sub>2</sub> O, CH <sub>4</sub> , NH <sub>3</sub> , COS, CO, NO, NO <sub>2</sub> , SO <sub>2</sub>	TGA100§	Campbell Scientific	closed path	Rinne et al. (2007)
Any two of: CO <sub>2</sub> , H <sub>2</sub> O, CH <sub>4</sub>	G2301-f¶	Picarro	closed path	Corbin et al. (2010; Crosson (2008); McDermitt et al. (2010)
CH <sub>4</sub> , CO <sub>2</sub> , H <sub>2</sub> O	Greenhouse Gas Analyzer	Los Gatos Research	closed path	Hsu et al. (2010)
CO <sub>2</sub> , H <sub>2</sub> O	EC150	Campbell Scientific	open path	#
CO <sub>2</sub> , H <sub>2</sub> O	EC155	Campbell Scientific	closed path	#

† These instruments are discussed in the Ameriflux methods (<http://public.ornl.gov/ameriflux/sop.shtml>).

‡ The earlier version of a closed-path analyzer from Li-Cor, the Li-6262, has also been widely used in the literature; it has since been discontinued.

§ Rinne et al. (2007) used a TGA100 which was replaced by the TGA100A and subsequently the TGA200, which is the currently available model.

¶ Older models from Picarro, Inc., have also been widely used.

# The Campbell Scientific EC150 and EC155 were released in the third quarter of 2010, so that at the time of writing, no studies have yet been published using the devices. It is expected, however, that this will be widely used in the future.

### Setup Considerations

For the mathematical algorithm behind eddy covariance to be valid, the surface must be horizontally homogeneous, so site selection is very important (Baldocchi, 2003). A flat terrain with acceptable fetch and an even canopy cover is essential for the measurements to be fully valid. If the site is not ideal, a footprint model will need to be used in post-processing (Schmid, 2002). Once an acceptable location is determined, the instrument setup should be twice the canopy height to be outside the roughness sublayer; additional height considerations are dependent on the chosen instruments (Foken, 2008). The gas sampling inlet should be downwind from the sonic anemometer path and the separation distance is dependent on the instrument and its height. Foken (2008) provided some good rule-of-thumb guidelines. In general, we recommend following the Ameriflux published standards for setup and calibration of instruments (Munger and Loescher, 2006). The sampling duration needs to be long enough to capture convective motion but not so long that the diurnal cycle will play a role (Baldocchi, 2003); the instrument selection will also limit this. Ideally a 10- to 20-Hz sampling rate should be used and 30-min fluxes will result. It is also important to follow the manufacturers' recommendations for calibration practices.

### Flux Calculation

Actual calculation of the flux should be done after the entire measurement campaign is completed. The calculation requires several steps. Some research groups have published open-source software for flux calculations (Table 22-2). Prepackaged commercial systems may also come with post-collection software options. If a new data processing routine is going to be developed, it should be compared with the "gold standard" data from the Ameriflux program for accuracy (<http://public.ornl.gov/ameriflux/sop.shtml>). Gold standards for both open- and closed-path systems are available. The general processes are presented here; for a more detailed description of data processing considerations, see Lee et al. (2004).

Data should first go through a quality control process to eliminate any periods of rain or instrument diagnostic flags that indicate abnormal functioning or adverse operating conditions. If gaps are large, gap-filling techniques can be used to fill in the missing data. The sonic anemometer data should undergo tilt correction—coordinate rotation routines to uphold the assumption of a zero-mean vertical wind component. The equations for tilt correction can be found in Wilczak

**Table 22-2.** Available Software programs for eddy covariance flux calculations.

Software	Source	Cost
EdiRe	Institute of Atmospheric and Environmental Sciences, School of Geosciences, University of Edinburgh	free
ECpack†	Meteorology and Air Quality Group, Wageningen University	free
TK2	Thomas Foken via Department of Micrometeorology, University of Bayreuth	free, registration required
Alteddy	Climate Xchange, Wageningen University	free

† van Dijk et al. (2004).

et al. (2001) and a review of the limitations can be found in Mahrt (2010). In addition, other data-cleaning techniques such as subtraction of the mean or detrending can be performed, although detrending is not always recommended in all situations (Baldocchi, 2003). Additional data processing techniques are discussed in Massman (2000).

### Limitations

While the eddy covariance method is widely used and a well-accepted technique, there are some limitations to the process of which a practitioner should be aware. First, it is only accurate under certain conditions (Baldocchi, 2003). Second, there is a potential for systematic errors in the energy balance closure (Twine et al., 2000) and a possibility of underestimation of nighttime fluxes in low wind speeds (Baldocchi, 2003; Twine et al., 2000), although more recent studies have shown good agreement between nighttime eddy covariance methods and chamber measurements (Law et al., 2001).

### SUMMARY

Chamber methods have made a significant contribution to the early assessment of global greenhouse inventories from various ecosystems. Recent advancements in various tower-based methods, especially improvement in gas detectors, makes them an important supplement to narrow the uncertainties in the greenhouse gas budget from ecosystems. Each method has its advantages and limitations. By comparing the measurements from these two methods, correction factors may be obtained to revise previous results.

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